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c0022 Treatment of flue-gas impurities for liquid absorbent-based postcombustion CO_2 capture processes

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s0010 22.1 Introduction

p0030 The first half of the 20th century witnessed the translation of initial concerns about power plant emissions into public pressure. This continued to build over subsequent decades in response to growing research data and awareness about the environmental and health impacts of emissions. The piecemeal development and implementation of Air Pollution Control Devices (APCDs) changed pace under incrementally stringent pollution control regulations in the last 25 years of the century. Advancements in APCDs accompanied their decreasing costs, in line with ever-tightening regulatory standards.

p0035 Of all the energy fuels, coal is of particular interest, owing to its abundance and ubiquitous worldwide presence, its 41% of world electricity production share, and its heterogeneous composition. Coal is projected to remain heavily mined, transported, traded, and consumed: most of it as thermal coal for power generation. The major producers of coal for local consumption and export (normally subbituminous thermal coals) are located in Indonesia, Australia, South Africa, and the United States. Low-
[AU3] rank coals (lignite or brown coal) are used only for local mine-mouth applications, such as heat or power generation. Lignite deposits are found in the European Union (Germany, Poland, Czech Republic, Greece, and Turkey); North America, across the boundary between Canada and the United States; the south of the United States; northern China; Russia; and the southern states of Australia. Several of these deposits contain more than 100 years' worth of proven reserves at current production rates.

p0040 Within any coal basin, there is considerable lateral and vertical variation in coal composition, making subsequent pollution control in combustion processes intrinsically challenging. The process of coal combustion generates flue gas, and untreated flue gases from the boiler are notorious for their levels of contaminants. The flue gases consist mainly of nitrogen (N_2), water vapor (H_2O), carbon dioxide (CO_2), and oxygen (O_2), with typical impurities including sulfur-containing compounds (SO_X), nitrogen oxide compounds (NO_X), carbon monoxide (CO), fly ash, soot (unburnt carbon), and trace elements. A sample of coal may contain up to 76 of the 90 naturally occurring elements in the periodic table (Schweinfurth, 2003), and as many

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as 120 different minerals (Nalbandian, 2012). A selected range of coal compositions is summarized in Table 22.1.

p0045 Flue-gas pretreatment is a critical step in conditioning flue gas to enable optimal performance of the postcombustion CO_2 capture (PCC) process. All PCC technologies are susceptible to detrimental impacts from untreated flue gas. For example, membrane processes do not perform optimally at high fly ash contents, especially when combined with high humidity. The same holds true for most adsorption or solid absorbent processes. Most liquid absorbent PCC processes—even aqueous amine absorbents, the most advanced technology for selective removal of CO_2—might be able to initially handle fly ash and moisture, but their long-term operation will be affected by flue-gas constituents.

p0050 Suboptimally conditioned flue gas accelerates the consumption of liquid absorbents, is likely to cause more corrosion, and increases the potential for environmental impacts through additional emissions. It also challenges process operations, causing problems such as foaming or local temperature runaways, and increases the discharge volumes of solid and slush waste streams while increasing maintenance requirements and strongly reducing the operation window. As a result, flue-gas treatment should be incorporated in a holistic approach to optimizing CO_2 capture.

p0055 This chapter introduces the different constituents of flue gases and the problems they are likely to cause. Individual flue-gas constituents are then discussed in light of current APCDs, such as deNOx units, electrostatic precipitators (ESP) or bag filters, and flue-gas desulfurization (FGD) units. We also investigate future additional pollution controls, such as deep-FGD, to allow for optimal operation and maintenance of PCC. The chapter concludes with an overview of recent advances in multicomponent removal strategies, including CO_2 capture.

s0015 *22.1.1 Technologies for treatment flue-gas impurities relevant to postcombustion CO_2 capture*

p0060 The treatment of flue-gas impurities has been addressed for more than a century. At one time, coal-fired power stations emitted large quantities of ash, soot, SO_X, and NO_X, as well as some mercury, and other trace elements. As a result, human respiratory systems were affected; nearby, freshly washed white sheets dried to shades of gray; fish built up high mercury levels; and acid rains slowly but steadily destroyed forests over decades of exposure. In modern power plants these issues have been successfully addressed. From about the 1950s, soot and fly ash were removed through filter bags and later, ESPs. In the 1970s, plant maintenance and acid rain drove the introduction of control measures for acid-related compounds, such as SO_X and NO_X. As a consequence, the concentrations of gaseous acid constituents decreased from many hundreds or thousands of ppm to under 50 ppm. A new ultra-supercritical power plant in China, built by Shenhua Guohua Electric Power, is reported to produce near-zero emissions for particulate matter (PM), sulfur dioxide (SO_2), and NO_X. A recent report concluded that the new plant was cleaner than a natural gas-fired power station, except for CO_2 emissions (Zhou, 2015).

t0010 **Table 22.1 Small selection of coals, highlighting the range of compositions**

Composition	Lignite			Bituminous		
	Australia[a]	United States[b]	China[c]	Australia[d]	United States[b]	China[c]
Proximate analysis (%)						
Moisture (ar)	60.6	35.88	2.6	3.3	2.33	5.19
Ash (db)	2.3	9.38	32.8	15.3	9.70	14.70
Volatile matter (daf)	50.7	28.15	25.84	28.1	39.19	24.28
Fixed carbon (daf)	47.2	26.59	38.76	56.6	48.78	55.83
Specific energy (GJ/t)	10.43	14.8	18.78	29.28	29.9	23.20
Ultimate analysis (daf) (%)						
Carbon	67.5	38.57	51.41	82.5	71.07	65.82
Oxygen	26.09	44.41	7.17	9.8	10.17	6.77
Hydrogen	4.9	6.59	3.19	5.10	5.07	3.00
Nitrogen	0.53	0.42	0.97	1.8	1.37	0.84
Sulfur	0.98	0.63	1.86	0.41	2.67	3.68
Chlorine	n.a.	n.a.	n.a.	n.a.	0.097	n.a.
Sodium in ash	2.6	n.a.	n.a.	n.a.	n.a.	n.a.
Ash analysis (%)						
SiO_2	16.4	22.00	51.16	56.1	55.83	33.33
Al_2O_3	3.4	20.38	30.15	32.6	34.27	19.98
Fe_2O_3	9.3	11.75	6.88	6.65	5.18	15.6
TiO_2	n.a.	0.50	1,37	1.09	1.71	1.10
P_2O_5	n.a.	n.a.	n.a.	0.10	0.23	n.a.
CaO	24.7	30.25	2.72	1.00	1.84	8.52
MgO	14.2	8.00	0.64	0.70	0.60	6.28
Na_2O	4.9	5.12	001	0.23	0.32	0.33
K_2O	0.3	1.38	0.52	0.63	1.53	0.8
SO_3	26.6	n.a.	2.74	n.a.	1.45	10.02
BaO	n.a.	0.50	n.a.	n.a.	n.a.	n.a.

ar, as received; *daf*, dry ash-free; *db*, dry basis; not applicable.
[a]Durie (1991).
[b]Senior et al. (1999).
[c]Wang et al. (2011).
[d]Huleatt (1991).

s0020 ### 22.1.2 Existing technologies

p0065 In flue gas, the principal species of concern are SO_2, NO_X, and PM, which are termed "criteria" air pollutants. APCDs are designed to reduce the emissions of these pollutants.

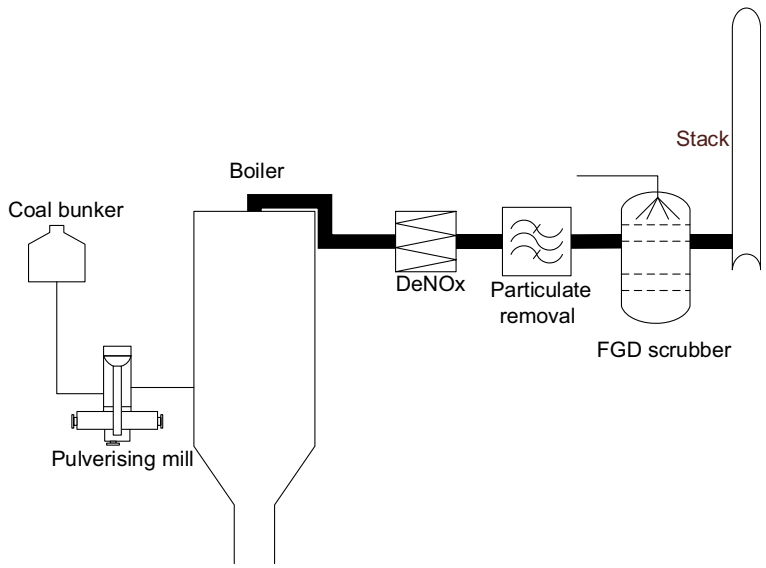
p0070 In practice, two or more technologies are installed to meet regulatory standards. They work in tandem to increase the overall removal capacity of specific pollutants. A typical basic train of emission control systems for NO_X, SO_X, and PM is depicted in Fig. 22.1.

p0075 Today's power plants incorporate a range of process unit operations to handle flue-gas impurities. A typical plant would have a NO_X treatment step, in which a noncatalytic nitrogen reduction step reduces NO_X to N_2 and water. A consequence of the deNOx process is that it typically adds ammonia (NH_3) to the flue-gas composition, which is a compound of concern. If Selective Catalytic Reactor technology is used for NO_X control, it also generates sulfur trioxide, thereby increasing its emission concentration (Carpenter, 2012).

p0080 In the next step, an ESP generates and collects particles. Active carbon can be added to capture organic volatiles and a baghouse filters particles. During the particulate capture process, sometimes sulfur trioxide (SO_3) is added to increase the overall removal efficiency, thereby increasing its concentration in the flue gas (Nicol, 2013).

p0085 Chlorides typically originate from salt layers that formed in the coal seam as seas dried up. Upon condensation, hydrochloric acid (HCl) in the flue gas could cause severe pit corrosion. Therefore, chloride ions are washed out with sulfur-loaded lime from the FGD unit. In the latter process step, limestone particles might be added to the flue-gas stream that might require clean up.

p0090 In some processes, another baghouse filter polishes the flue gas, or a caustic wash is added to further decrease SO_2 levels before the gas is emitted to the atmosphere. A fan



f0010 **Figure 22.1** Typical emission control systems on a coal power plant.

is required to maintain proper dispersion, because cooling, removal of water, and pressure drop through the processes all considerably reduce flue-gas buoyancy.

p0095 Some forms of APCD are listed in Table 22.2.

s0025 22.1.3 Considerations for flue-gas treatment to optimize PCC

p0100 A PCC plant is built downstream of pollution-control technologies, to ensure maximum removal of contaminants from the flue gas before it enters the PCC section. To ensure smooth operation, such technologies are also needed to condition the flue gas before it enters the PCC plant (Azzi et al., 2013). The impurities must be removed between the APCD and the PCC plant to ensure that they do not contaminate the CO_2 stream destined for geological storage, because they may affect the transport system or interfere with the underground reservoir. Smooth operations of the PCC plant would actually dictate cleaner flue gas than currently permitted by emission legislations around the globe (Table 22.3), and result in lower emissions. In contrast, PCC-equipped power plants, with their lower overall efficiency and additional waste streams, generate more waste than a similar-sized conventional power plant (Azzi et al., 2013).

p0105 Liquid absorbents used for CO_2 removal also capture the remaining impurities in the flue gas (<2 ppm for SO_2 or nitrogen dioxide [NO_2]), thereby further reducing their emissions to the atmosphere. Although this is a desirable effect, it results in the accumulation of these compounds and their reaction products within the PCC plant. The impact of these substances on the liquid absorbent and the PCC plant's operations is of concern. In cases in which the harm of an incoming flue-gas impurity outweighs the benefit of its capture, it needs to be removed before entering the capture plant. Acid gases such as NO_X and SO_X pose the biggest challenge; they compete with CO_2 for the liquid absorbent, degrade the absorbent, and create heat-stable salts. Trace elements are the second-largest challenge. Amine scrubbing removes most of them, but they eventually end up in the liquid absorbent, in which they cause operational issues. Overcoming these issues requires either modification of existing APCDs in the operational power plant, or installation of new technologies to achieve the desired level of impurity concentration. However, the selection of a technology to tackle a specific impurity depends on many variables, including

u0025 • fuel type
u0030 • mechanical factors
u0035 ◦ power plant design
u0040 ◦ PCC plant design
u0045 ◦ capacity
u0050 ◦ footprint
u0055 • power plant location/geography
u0060 ◦ access to local market (eg, byproduct sale)
u0065 ◦ access to resources (eg, seawater for desulfurization)
u0070 • impurity concentration and required reduction
u0075 • cost factors
u0080 ◦ power plant's economic life
u0085 ◦ capital cost
u0090 ◦ operational cost
u0095 ◦ discount rates
u0100 ◦ inflation rates.

t0015 **Table 22.2 Emission control systems used in the US coal-fired boilers in 2010, for boilers >25 MW (Amar et al., 2010)**

[AU4]

Emission control system	Bituminous	Subbituminous	Lignite	Other (waste coal/petroleum coke [petcoke])	Total
Selective catalytic reduction	208	84	2	1	295
Selective noncatalytic reduction	146	22	2	15	185
Cold-side electrostatic precipitator	454	257	17	1	729
Hot-side electrostatic precipitator	78	23	0	0	101
Fabric filters	120	112	12	31	275
Other particulate matter control	16	14	1	0	31
Wet flue-gas desulfurization	255	89	16	1	361
Spray dryer	54	31	3	0	88
Fluidized bed flue-gas desulfurization	20	4	7	29	60
Dry sorbent injection flue-gas desulfurization	29	7	1	0	37

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t0020 **Table 22.3 Emission limits for new power plants (μg/m^3) versus flue gas versus postcombustion CO_2 capture (PCC) plant requirements**

Flue-gas component	Flue-gas composition				Emission limits				
	Lignite		Black coal						
	AU[a]	USA[b]	AU[c]	USA[b]	AU[d]	USA[e]	China[f]	EU[g]	PCC plant
NO_X	150−250 ppm	>300 ppm	150 ppm	>300 ppm	500−800	134−515	100−200	150−300	5−40 (NO_2) ppm
SO_X	120−200 ppm	840 ppm	200 ppm	1900 ppm	200[h]	186−515	50−400	150−400	3−30 (SO_2) ppm
PM	n.a.	n.a.	n.a.	n.a.	80	18−52	20−30	20−30	n.a.
Hg	n.a.	n.a.	n.a.	n.a.	n.a.	0.0003−0.005	0.03	0.05	n.a.
HCl	n.a.	5 ppm	n.a.	60	n.a.	n.a.	n.a.	n.a.	n.a.
H_2O (%)	20−23	14.7	8	8	n.a.	n.a.	n.a.	n.a.	n.a.
CO_2 (%)	10−11	14	10	14	n.a.	n.a.	n.a.	n.a.	n.a.
O_2 (%)	4−5	3.0	6	>3.0	n.a.	n.a.	n.a.	n.a.	n.a.
N_2 (%)	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

n.a., not applicable.
[a]Artanto et al. (2012).
[b]Senior et al. (1999).
[c]Cousins et al. (2012).
[d]IEA Australia (2015).
[e]IEA USA (2015).
[f]Minchener (2012).
[g]EU Journal (2010).
[h]Expressed as SO_3.

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p0190 Based on the aforementioned factors, or any other relevant criteria, a technology is selected as either "best available technology" or "maximum achievable control technology." These terms are usually synonymously used to define the most appropriate technology for reducing emissions of a certain pollutant.

p0195 The following sections discuss technologies that address particular impurities. Oxygen is not discussed as it is not an impurity as such, and we are not aware of any process in which oxygen is completely removed before the PCC process. The aim of oxygen removal could be to reduce corrosion and degradation rates of liquid absorbents, hence reducing the requirement for oxygen-scavengers in the process.

s0030 22.2 NO_X control

p0200 Nitrogen content in coal ranges between 0.5% and 2%, contributing to the formation of nearly 80% of the nitric oxide (NO) and NO_2 found in flue gas. Collectively, these compounds are called NO_X. Some of the nitrogen in coal is converted into N_2 when coal is combusted; a small amount of nitrous oxide (N_2O) is also formed. N_2O is a potent greenhouse gas with 298 times the global warming potential of CO_2. The final composition of nitrogen compounds in the flue gas depends upon the intermediate radical reactions, temperature, and oxygen availability in the reaction regions.

p0205 Despite the health and environmental impacts of NO_X being known for decades, only stringent government regulations forced NO_X control technological innovation (Yeh et al., 2005), thereby reducing the capital and operating costs of the technology (Fig. 22.2). Primary measures, otherwise known as combustion modifications (Nalbandian, 2009), were the first methods developed to control NO_X emissions in the 1970s. In the 1980s, the NO_X reduction (deNOx) technologies of selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR) were developed.

p0210 The following sections discuss NO_X impacts and technologies for emission reduction. The NO_X control technologies included are focused on commercially mature systems, with a slight focus toward PCC plants.

s0035 *22.2.1 NO_X emission effects*

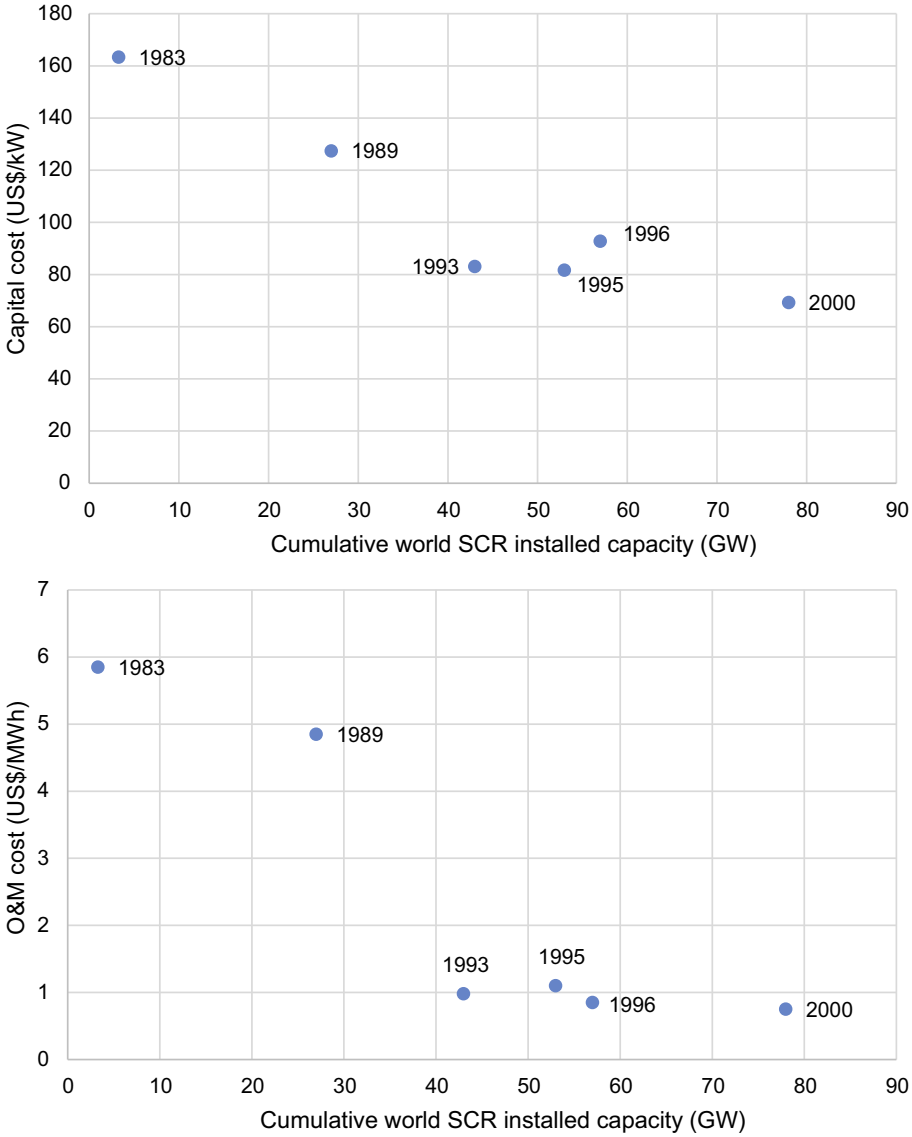
p0215 The environmental impacts of NO_X are well documented. NO_X are major contributors to the formation of acid rain in the atmosphere via the following chemical reactions.

p0220 Nitrogen dioxide reacts with water:

$$2NO_2 + H_2O \rightarrow HNO_2 + HNO_3$$

p0225 Nitrous acid then decomposes as follows:

$$3HNO_2 \rightarrow HNO_3 + 2NO + H_2O$$



f0015 **Figure 22.2** (Top) Capital costs and (bottom) operational and maintenance (O&M) costs of selective catalytic reduction (SCR) deNOx systems, 1983–2000 (Yeh et al., 2005).

p0230 Nitric oxide oxidizes to form nitrogen dioxide and reacts with water, ultimately forming nitric acid:

$$4NO + 3O_2 + 2H_2O \rightarrow 4HNO_3$$

p0235 In more simple terms, the overall reaction is expressed as ($NO_X + H_2O \rightarrow HNO_3$).

p0240 NO_X play a key part in the photochemically induced catalytic production of ozone, which results in summer smog (Richter et al., 2005), and in the second half of the 20th century increased the global tropospheric ozone levels (Volz and Kley, 1988). NO_X also contribute to eutrophication of terrestrial and aquatic ecosystems. NO_2 is a major air pollutant, and its concentration is substantially increasing over the industrial areas of China (Richter et al., 2005). Stringent NO_X emission limits have significantly reduced NO_2 concentrations over some areas of Europe and the United States (Richter et al., 2005). At local levels, at least, NO_2 increases radiative forcing effects due to the absorption of downward-propagating visible light (Solomon et al., 1999).

p0245 The health impacts of NO_X emissions have also been well researched. Both short and long-term exposure causes respiratory health issues in humans.

p0250 PCC plant operation requires minimal NO_2 contamination in the flue-gas stream, whereas NO is an inert species in a PCC plant. In a liquid absorbent-based PCC process, NO_2 is an acid gas that reacts with the amine, generating heat-stable salts, and causing degradation and loss of amine. Process vendors specify NO_2 limits for the incoming flue-gas stream (eg, the Fluor Econamine process has an upper limit of 20 ppm (Adams, 2010) for NO_2). Some NO_X generate nitrosamine compounds after reacting with amine solvents (Fostås et al., 2011; Jackson and Attalla, 2011), and N_2O is soluble in aqueous amines (Ma'mun and Svendsen, 2009).

s0040 *22.2.2 Primary measures (combustion modifications)*

p0255 Primary measures reduce the temperature, oxygen, or residence time at the peak combustion temperature, all of which favor N_2 formation over NO_X. These technologies have been in use for decades, with low-NO_X burners and over-fire air systems retaining the biggest market share.

p0260 In pulverized-coal combustion, fluidized-bed combustion and cofiring (with biomass), the primary measures are implemented as a first step toward controlling NO_X emissions (Nalbandian, 2009). Supplementing these with deNOx technologies further reduces these emissions. Although biomass cofiring is used as a NO_X reduction method, NO_X emissions can both increase and decrease with cofiring (Nalbandian, 2009), depending upon biomass composition.

p0265 Five main primary measures for NO_X reduction are commonly applied:

o0010 **1.** process optimization (reduces NO_X by up to 20%)

o0015 **2.** low-NO_X burners, in which the air and fuel mixing at each burner are controlled to minimize NO_X formation (reduces NO_X by 60%)

o0020 **3.** air-staging method (over-fire air system), in which separated combustion air in the furnace achieves complete combustion, thereby encouraging N_2 formation over NO_X (reduces NO_X by 50–70%)

o0025 **4.** fuel staging or reburning, which involves consuming the oxygen in the air during the burning process, thereby converting NO_X to N_2 (reduces NO_X by 50–70%)

o0030 **5.** flue-gas recirculation (reduces NO_X by 50–80%).

s0045 22.2.3 Nitrogen reduction (DeNOx) technologies

p0295 DeNOx technologies can be used independently or in combination with the primary measures to reduce NO_X emissions. These technologies principally work by injecting ammonia, urea, or other chemicals into the flue gas to reach the NO_X, thereby reducing them to N_2 and water. SCR and SNCR systems are the two main deNOx technologies, with the main difference between the two being the application or absence of the catalyst. In the presence of the catalyst, the reaction takes place from around 160−350°C, whereas in its absence, the temperature requirement increases to around 1000°C.

p0300 SCR technology retains the biggest deNOx market share in coal-fired power plants. The system operates by injecting NH_3 into the flue gas as it passes over a catalyst. The NH_3 acts as a reducing agent. SCR removes nearly 95% of the NO_X in the flue gas (both NO and NO_2) while maintaining NH_3 slip to 5 ppm or less.

p0305 Catalysts can be made from a variety of material, including titanium oxide, zeolite, iron oxide, and activated carbon. The final catalyst composition may also include many active metals and support materials. A mixture of vanadium (active catalyst) and titanium (used to disperse and support the vanadium) is mostly used in coal-fired power plants. Higher vanadium concentrations not only result in greater NO_X reduction but also oxidize SO_2 to SO_3. The total quantity of SO_3 generated in the system equals the amount produced in the boiler, thereby potentially doubling the concentration in the flue gas (Nicol, 2013). SO_3 can further react with unreacted NH_3 to form either submicron ammonium sulfates, which are difficult to capture; or low-melting-point sulfates, such as ammonium bisulfate (NH_4HSO_4) (Nicol, 2013). Ammonium bisulfate increases particulate cohesiveness, creating issues in particulate removal downstream. In the presence of NH_3, SO_3 formation increases greatly at temperatures above 370°C. This is solved by limiting temperatures to below 370°C in applications using NH_3 as a reagent.

p0310 Due to SO_3 being acidic in nature, it is important to limit its formation in PCC applications. It degrades the liquid absorbent in the PCC plant, and tends to combine with the absorbent, forming aerosols that are difficult to control (Azzi et al., 2014). This subsequently gives rise to solvent loss and other plant issues. SO_3 removal from flue gas is discussed in Section 22.4.

p0315 Pilot and demonstration test results have indicated that the vanadia−titania catalyst used in SCRs can oxidize elemental mercury (Hg^0) (Electric Power Research Institute [**AU5**] (EPRI, 2002a,b)) to Hg^{2+} (Wilcox et al., 2012). This improves mercury capture in the particulate control device (discussed in Section 22.5). However, in the absence of HCl, mercury oxidation is negligible.

p0320 SNCR systems are based on injecting urea or anhydrous gaseous NH_3 into the hot flue gas to react with and convert NO_X to N_2 and water vapor. SNCR systems operate at high temperatures, with exit temperatures of around 1000°C. They can reduce emissions from coal-fired power plants by up to 75%, but are usually only intended for small to medium boilers. Although no SO_2 is oxidized to SO_3 in the process, the technology is still plagued by the issue of unreacted NH_3 reacting with already available SO_3 to form ammonium bisulfate. Ammonium bisulfate precipitates at air heater operating temperatures, leading to air heater fouling and plugging. High NH_3 slip rates also

cause NH_3 odors around the plant, whereas absorption of NH_3 onto fly ash makes its disposal or reuse troublesome. The SNCR reagent incurs the main operational costs.

s0050 22.2.4 Other technologies and synopsis

p0325 Many NO_X control technologies under development or in the demonstration stages aim to reduce costs or increase efficiency. Noteworthy are hybrid SNCR/SCR technology; LoTOx technology, which has been demonstrated on a number of industrial plants; the ECOTUBE system; and the NOxStar control system (Nalbandian, 2009).

p0330 Waste amine from PCC plants also has deNOx capacity. Studies have indicated that degraded liquid amine waste can significantly reduce NO_X emissions in flue gas (Botheju et al., 2012, 2013); however, more research is needed in this area. Aqueous solutions of $KMnO_4$ (Brogren et al., 1997) and $KMnO_4/(NH_4)_2CO_3$ (Pan et al., 2013) for NO absorption have also been investigated.

p0335 Table 22.4 gives an overview of the various NO_X control systems available.

s0055 22.3 Particulate matter control

p0340 Mineral matter in coal ends up as either fly ash or bottom ash after combustion. Up to 80−90% of the mineral matter may be emitted as fly ash with the flue gas. The composition, mass, and shape of the particulates in the fly ash depend upon the coal type, power plant design, and its operating parameters.

p0345 ESPs and fabric filters (FFs) are the two main technologies used to remove most of the fly ash before it reaches the stack chimneys. Despite ESPs removing up to 99.9% [AU6] (by mass) of particulates (Azzi et al., 2013) and FFs removing up to 99.95% (Nicol, 2013), certain portions of fine material and associated trace elements still pass through. The fly ash is recovered from ESPs and FFs, and if it meets the quality parameters, it is sold for use in concrete, cement, grouting, or niche products such as geopolymers. Wet scrubbers and cyclones are two other technologies for particulate control, but are not popular. Cyclones have inadequate collection efficiency at reasonable pressure drops (Nicol, 2013), whereas wet scrubbers need wastewater treatment systems, which have high levels of water and energy consumption.

s0060 22.3.1 Particulate emission effects

p0350 The health impact of airborne particulates relates to adverse heart and lung effects. Particles of less than 10 μm (PM_{10}) can enter into the respiratory tract, whereas particles less than 2.5 μm ($PM_{2.5}$) can cross into the bloodstream. Many studies worldwide have identified an association between long-term exposure to $PM_{2.5}$ and increased mortality risk (Brook et al., 2010; Kappos et al., 2004; Pope and Dockery, 2006). Increasing concern related to the fine PM emission is driving progressively stringent emission standards.

t0025 **Table 22.4 Overview of NO_X control systems**

Technology	Max removal efficiency (%)	Commercial availability	Reagent used	Retro-fit suitable	Multiproduct removal
Primary measures					
Process optimization	20	Y	N	Y	N
Low NO_X burner	60	Y	N	Y	N
Over fire air (OFA)	30	Y	N	a	N
LNB + OFA	60	Y	N	Y	N
Fuel staging	60	Y	N	a	N
FGR	80	Y	N	N	N
DeNOX technology					
SCR	95	Y	Y	Y	Y
SNCR	75	Y	Y	Y	N
Primary measures with DeNOx					
LNB with SNCR	80	Y	Y	Y	N
LNB + OFA + SCR	95	Y	Y	Y	Y

FGR, flue-gas recirculation; *LNB*, low-NO_X burner; *OFA*, Over-fire air; *SCR*, selective catalytic reduction; *SNCR*, selective noncatalytic reduction; *Y*, yes; *N*, no; *H*, high; *L*, low.
[a]Retrofitting requires sufficient burner height.

p0355 PCC plants may face operational problems due to PM, such as equipment blockage or foaming of the liquid absorbent (Iijima et al., 2007). A PCC plant works with saturated gases, which can lead to wet deposits of ash in pipe work. Due to the cementitious nature of some ashes, they can form very hard and stable deposits or blockages in the system. Liquid absorbents are recycled in a PCC plant. Therefore, when ash enters the liquid circuits, particulates (even at low concentrations) would accumulate over time: first in the liquid absorbent and then in the reclaimer waste. The combination of corrosion and fly ash will accelerate blockage as well as breakdown reactions of liquid absorbents, causing further corrosion while possibly increasing emissions and waste streams. The removal of fly ash is therefore desirable to avoid PCC-related operational issues.

s0065 22.3.2 Electrostatic precipitators

p0360 An ESP uses a discharge electrode to negatively charge the PM, collecting the particles on a positively charged plate electrode. For continuous operation, the accumulated particles must be removed either by rapping using a tumbling hammer or magnetic impact (dry ESP), or by water wash (wet ESP).

p0365 Conventional ESPs have a low $PM_{2.5}$ collection efficiency. This can be overcome with the use of an agglomerator, which binds fine particulates into larger particulates using various chemical and physical techniques. The larger particulates are then efficiently collected by the ESP.

p0370 If the fly ash resistivity is too high, the collection efficiency decreases. In this case, fly ash resistivity is lowered by humidification or by adding flue-gas conditioning chemicals, such as SO_3, NH_3, and proprietary chemicals. Several flue-gas conditioning system designs are available, all of which increase the operating cost. SO_3 is a popular option; it is highly conductive, and lowers fly ash resistivity by dissolving into water on particulates, forming sulfites. However, high SO_3 concentrations in the flue gas increase stack opacity, fouling, and corrosion. In a PCC plant, SO_3 degrades the liquid absorbent, necessitating either its complete removal or maximum reduction. SO_3 is also notorious for creating aerosols that pass through wash systems designed to prevent emissions from PCC operations.

s0070 22.3.2.1 Wet and dry electrostatic precipitators

p0375 In wet ESPs, the accumulated particulates on the electrodes are continuously washed with water spray nozzles. Wet ESP has the following advantages over dry ESP:

u0105 • The humidity lowers the flue-gas temperature, thereby increasing the specific collection area and lowering fly ash resistivity.

u0110 • Water cleaning replaces rapping, which eliminates the emission peaks created during rapping.

u0115 • A higher corona power can be used to increase charging.

u0120 • The collection efficiencies of PM_{10} and $PM_{2.5}$, sulfuric acid, and other soluble acid aerosols are higher.

u0125 • It can be integrated with various scrubbing techniques, such as wet FGD, paving the way for multipollutant control (Stultz and Kitto, 2005).

p0405 Drawbacks of wet ESP include its high water consumption and excess wastewater generation, which requires costly and complicated treatment systems. The flue gas exiting the wet ESP is cooler than that exiting cold-side dry ESP, and requires heating for some gas cleanup technologies downstream. Plus, under certain parameters, the SO_3 mist attains a pH of 0.5, which is very corrosive. ESP equipment must therefore be made from expensive, corrosion-resistant material.

p0410 Advancements in ESP systems are continuing. Noteworthy new benchmark technologies include Hitachi Plant Technologies (Japan) moving electrode electrostatic precipitator, electromechanical double-zone ESP, ion-blast ESP, and cross-flow ESP (Nicol, 2013).

s0075 22.3.3 Fabric filters

p0415 FFs, or baghouse filters, operate on relatively simple processes compared with ESP and have better particulate removal efficiency. They work on the principle of vacuum cleaners (Nicol, 2013); the flue gas passes through a filter, which rejects particles that are too large to pass through. Fly ash is collected by the bag/filter itself, or by the accumulated fly ash on the filter (called filter cake).

p0420 As mentioned previously, FFs have up to 99.95% overall PM removal efficiency (Nicol, 2013). Unlike ESPs, in which resistivity is an important factor, FF collection efficiency is not directly related to resistivity. This makes the technology suitable for highly resistive fly ashes. The pressure drop across an FF is much greater than that across an ESP, which is compensated for by a high-powered induced fan. Filter cake management is the most important factor for optimal performance (Nicol, 2013). When a certain amount of filter cake builds up, it must be removed before air pressure buildup breaks the filter.

p0425 [AU7] Improvements such as microprocessor-based control, flow control devices, cohesiveness control, and conditioning agents can further increase fly ash collection efficiency. Elemental S, NH_3, and SO_3 are the usual candidates for conditioning chemicals. Overloading the primary filter layer with cohesive particulates results in a large pressure drop. Particulate cohesiveness increases with ammonium bisulfate levels (from SCR NH_3 slip upstream) and activated carbon sorbents. Lower-cohesive particulates create pores in the bag, causing PM to bleed through. FFs are not used in high-moisture flue gases, such as those originating from lignites, because the filters would easily clog.

s0080 22.4 SO_X emission control

p0430 Coal naturally contains a small amount of sulfur, in the form of organically bound sulfur (as part of coal's molecular structure), fine pyritic particles (sulfide minerals), and sulfate minerals. Sulfate minerals represent a small portion of total sulfur content, and do not contribute to emissions.

p0435 During coal combustion, organic and pyritic sulfur are oxidized to form SO_2 in the boiler. In the presence of coal's catalytic metals, a small portion of the SO_2 is further oxidized to SO_3. The amount of SO_2 and SO_3 formed depends upon the coal composition, boiler design, and combustion parameters. Flue-gas processing design also dictates the final concentration of SO_2 and SO_3 in the flue gas. For example, SCR deNOx leads to catalytic oxidation of SO_2, and even doubles the amount of SO_3 in the flue gas in some instances. In addition, SO_3 is sometimes used as a flue-gas conditioning chemical in ESP to reduce fly ash resistivity. This increases the total SO_3 concentration requiring treatment in the FGD unit, which captures SO_X from the flue gas after combustion process.

p0440 Ideally, precombustion desulfurization would prevent SO_X emissions. Being a new area of research, much effort is required before such systems reach commercial maturity. Current FGD units typically used at power stations generally do not reduce SO_X concentration to the levels required for a PCC plant.

p0445 For PCC technology using monoethanolamine (MEA)-based solvent, SO_2 concentrations below 10 ppm would be required (Adams, 2010; Rao and Rubin, 2002) to avoid excessive solvent loss. The limit would be even lower for other, more expensive
[**AU8**] amines (Endo et al., 2011). FGD processes such as those used in Shell's Cansolv® process achieve this level of removal efficiency, whereas alkaline salt solutions can meet this rigid specification if used in a spray scrubber (Adams, 2010) or a packed column (Artanto et al., 2012). Other than SO_X removal efficiency, the selection of an FGD technology depends on factors such as its water usage, cost, commercial status, size and location of the plant, and duration of the plant shutdown if retrofitting.

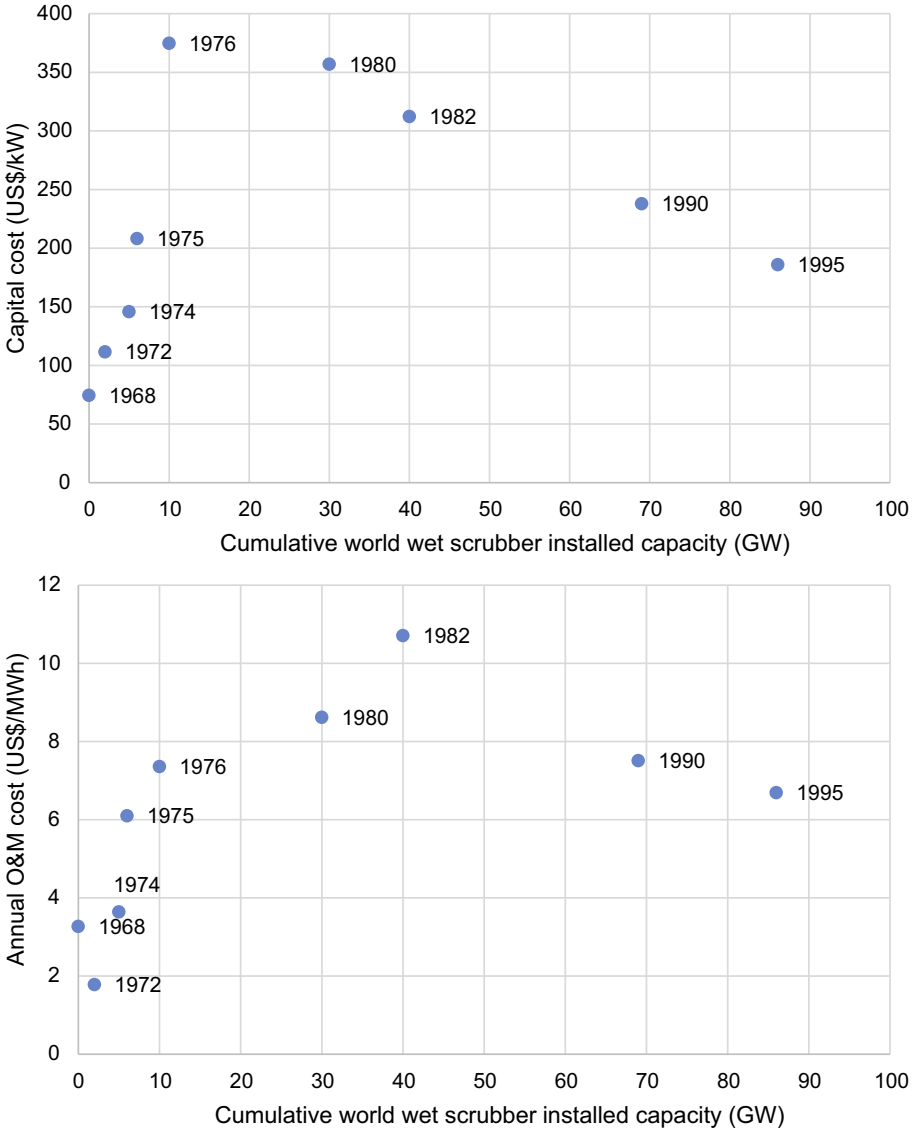
p0450 The FGD concept germinated in England around 1850. The primary research was related to the quest to uncover the fundamental physical properties of substances (Biondo and Marten, 1977). In the 1860s, experiments were performed on SO_2 absorp-
[**AU9**] tion in water (Sims, 1862; Watts, 1864). The role of catalysts in SO_2 absorption was discovered shortly thereafter (Biondo and Marten, 1977). Commercially usable sulfur recovery from coal flue gases became the impetus for FGD advancements at the turn of the century (Biondo and Marten, 1977), and the first large-scale FGD units began commercial operation in the 1930s in England. In light of SO_2 control regulations, a host of FGD technologies were commercially implemented in the United States and Japan in the 1970s (SEL, 1973), with wet scrubbing being the foremost.

p0455 Despite an increase in the total installed capacity of FGD technologies, the capital and O&M costs continued to climb through the 1970s, beginning to fall in the 1980s and continuing through the 1990s (Fig. 22.3) (Rubin et al., 2006). Although several factors led to the cost increases in the 1970s, shortfalls in performance and reliability were the predominant causes (Rubin et al., 2006).

p0460 Wet scrubbing remains the main commercial FGD technology, with over 84% market share, followed by semidry (>6.1%) and dry processes (1.6%) (Zhu, 2010). As is apparent, the three technologies differ according to their usage of water. The other difference lies in their sorbent regeneration capacity (regenerative processes) and waste/byproduct formation. In regenerative processes, the reagent used for SO_2 removal is regenerated and reused in the scrubber.

p0465 In the majority of wet scrubber technologies, a particulate control system is installed upstream of the wet SO_2 scrubber. However, in dry scrubbing technology, the SO_2 is first reacted with the sorbent, and then the flue gas passes through the particulate control system. Attempts to remove particulates and SO_2 in the same step lead to maintenance issues and low removal efficiencies. The basic operation of almost all the commercial FGD technologies is based on the acidic nature of SO_X, which are removed by reacting with a suitable alkaline compound. This is commonly limestone (calcium carbonate), which is cheap and abundantly available.

p0470 In the carbon capture context, the necessity of very low SO_X concentration requires either modifying the existing SO_X control technologies or replacing them with new systems that achieve the required SO_X concentration. In some scenarios, such as when the capture plant would be retrofitted, the two options might not be suitable; then SO_2 Polishing is utilized. SO_2 Polishing or Deep FGD refers to the installation of an additional technology/system downstream of the existing SO_X control technology that achieves the required removal rate. In the polishing stage a scrubbing solution



f0020 **Figure 22.3** (Top) Capital costs and (bottom) operational & maintenance (O&M) costs of wet limestone flue-gas desulfurization systems (Rubin et al., 2006).

with a pH of approximately seven is usually utilized, which has the capacity to reduce the SO_2 concentration to 1 ppm (vol) or less (Brady, 1987). The operational cost of a polisher is designed to be less than the additional cost of the MEA consumption due to SO_2-related losses (Adams et al., 2009).

p0475 Table 22.5 compares the main FGD technologies.

t0030 **Table 22.5 Overview of flue-gas desulfurization (FGD) technologies**

Technology	Max removal efficiency (%)	Max sulfur concentration	Commercial availability	Regeneration needed	By-prodcut	Retro-fit	Multiproduct removal	Capital cost	O&M cost
Wet FGD									
Limestone/Lime	98	H	Y	N	Y	n.a.	n.a.	H	L
Seawater	99		Y	N	N	n.a.	n.a.	L	L
Ammonia FGD	98	H	Y	N	Y	n.a.	n.a.	H	L
Wellman−Lord	95	H	Y	Y	n.a.	n.a.	n.a.	H	L
Mg(OH)2	98	n.a.	Y	Y	Y	n.a.	Y	L	H
Semidry									
Spray dry	95	M	Y	N	Y	Y	n.a.	L	H
Duct spray	75	n.a.	N	n.a.		Y	n.a.	L	n.a.
Dry FGD									
Circulating fluid bed	99	H	Y	n.a.	N	Y	n.a.	L	H
Furnace sorbent	80	n.a.	Y	n.a.	n.a.	Y	n.a.	L	H
Sodium bicarbonate	70	n.a.	Y	n.a.	n.a.	n.a.	n.a.	L	H
Metal oxides	90	n.a.	N	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Activated carbon	98	n.a.	n.a.	n.a.	n.a.	n.a.	Y	n.a.	n.a.
Electron beam	95	n.a.	n.a.	n.a.	Y	n.a.	Y	H	M

H, high; *M*, medium; *L*, low; *N*, no; *Y*, yes; n.a., not applicable.

s0085 22.4.1 SO_X emission effects

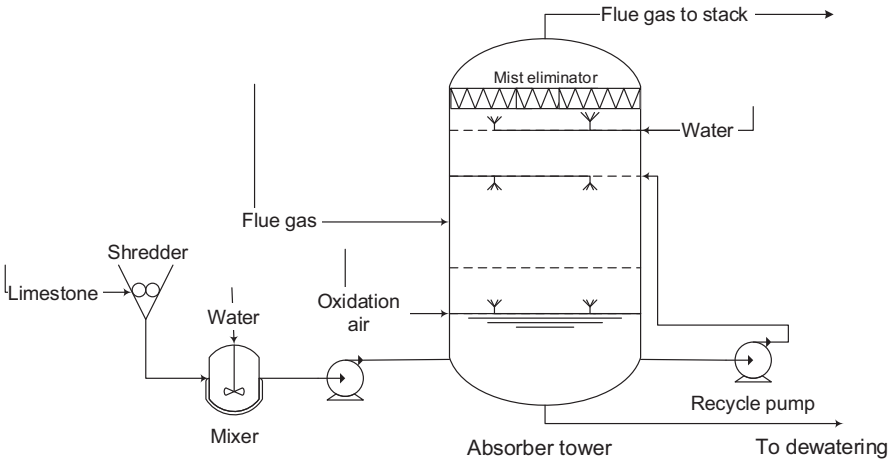
p0480 The environmental impact of SO_X emissions differs between SO_2 and SO_3. SO_2 generates acid rain and urban smog, and is hazardous to health. High concentrations of SO_3 form sulfuric acid, which causes plant equipment corrosion and fouling, and plume opacity.

p0485 SO_X also cause operational issues in PCC plants. They react with amine absorbents, forming heat-stable salts; reduce CO_2 absorption capacity; and increase absorbent makeup rates. This requires the removal of (or substantial decrease in) SO_X concentration in the flue gas before entering the PCC plant.

p0490 Coal-fired power plant flue-gas SO_2 concentrations may exceed 3000 ppm (Adams, 2010); however, the concentrations of SO_2 and SO_3 vary widely. For example, Powder River Basin coal in the United States produces virtually no SO_3, because its highly alkaline ash removes the bulk of the generated SO_3 (EPRI, 2007). Australian coal, on the other hand, produces around 200−300 ppm SO_2 (Artanto et al., 2012; Davis and Reddan, 2012). Measurements in a New South Wales power station showed that SO_3 represented only about 2% of the total sulfur in the flue gas (Davis and Reddan, 2012). In support of EPRI's Power River Basin findings, the SO_3 concentration was below the detection limit at a power station in Victoria (Australia), because Victorian brown coal also has highly alkaline ash (Artanto et al., 2012).

s0090 22.4.2 Wet scrubbers

p0495 In wet scrubbers (Fig. 22.4), the SO_2 is dissolved from the flue gas into water. The resulting liquid is collected and disposed of or recycled after separating the dissolved gas/particles. The SO_2 absorption rate is improved by using a cheap alkali agent, such as lime or limestone, in the liquid. Sodium-based solutions have also been also developed. They provide better SO_2 solubility and less scaling than lime or limestone, but are much more expensive. Power plants located near the ocean can use seawater as an



f0025 **Figure 22.4** Limestone wet scrubber system.

FGD reagent. Seawater is naturally alkaline, with a pH value of 7.6 to 8.4, and can absorb and neutralize acidic SO_2 in the flue gas.

p0500 The NH_3 scrubber process is similar to limestone−gypsum process, but uses a different reagent. Aqueous ammonia-based scrubbing can remove up to 98% of SO_2 in the flue gas, even if there is an extremely high-sulfur content. There is growing interest in developing NH_3-based scrubbing systems that remove SO_2 and CO_2 simultaneously. A magnesium hydroxide reagent-based scrubbing process removes SO_2 while simultaneously removing particulates in a single tower, which eliminates the need for a separate dust collection system.

p0505 The flue gas that leaves the wet scrubber still contains some SO_2 and water, which combination is highly corrosive. This requires the decision to either reheat the gases to above the dew point, or use equipment that can withstand corrosion (Makansi, 1982).

p0510 There are five main wet scrubbing technologies:

u0130 • limestone/lime gypsum process
u0135 • seawater scrubbing
u0140 • ammonia scrubbing
u0145 • Wellman−Lord process
u0150 • magnesium oxide process.

p0540 There are also four main wet scrubber designs (Takeshita and Soud, 1993), which are spray-tower scrubber, packed tower, jet-bubbling reactor, and double-loop reactor. Because of the issues of plugging, erosion, and scaling, which affect scrubber absorption efficiency and dependability, the simplest design of a spray tower is preferred.

s0095 *22.4.2.1 Strengths and limitations*

p0545 Wet scrubbers require a large space, and have high capital (Mochida et al., 2000) and operational costs. Additionally, they require large amounts of water and energy. Wet scrubbing is desirable for high SO_X concentrations, because of the high rate of limestone use. However, the system does not completely remove SO_2, with usually around 50 ppm SO_2 remaining in the flue gas (Mochida et al., 2000). Wet FGD also scrubs out approximately 60% of particulates from the ESP outlet, of which most are $PM_{2.5}$ (Nicol, 2013).

s0100 *22.4.3 Semidry scrubbers*

p0550 Semidry scrubbers are among the simplest designs and the technology is typically categorized into:

u0155 • spray dry scrubbers (spray dry absorber)
u0160 • duct spray dry process
u0165 • circulating dry scrubbers.

p0570 Semidry scrubbers consume 60% less water than the conventional wet scrubbers (Carpenter, 2012). Calcium-based reagent (calcium hydroxide) is typically used as the sorbent either in an aqueous slurry form or as a dry powder, and a mixed solid waste is generated for disposal (Zhu, 2010).

p0575 Spray dry scrubbers remain the second most widely installed system after wet scrubbers in the coal-fired power plants with a total installed capacity of around 25 GWe by the end of 2008 (Zhu, 2010). SO_2 removal efficiencies reach 90–95%, depending upon the sulfur content of coal (Carpenter, 2012). Spray dry scrubbers are usually installed on small- to medium-sized (~450 MW) units (Carpenter, 2012) combusting low to medium sulfur (2% S) coals (Moss, 2010).

s0105 *22.4.4 Dry scrubbers*

p0580 Activated carbon originated as a sorbent material for human protection against chemical exposure during World War I. It is now found in numerous industrial applications. In the 1960s, activated carbon was shown to be an effective dry desulfurization technology, via SO_2 adsorption into its pores in the presence of oxygen and water vapor at about 100°C (Tamura et al., 1968). SO_2 is adsorbed as H_2SO_4 into the activated carbon, and must be removed to regenerate the activated carbon's adsorption ability. Activated carbon can also remove multiple contaminants from flue gas.

p0585 In a typical electron-beam dry scrubbing process, the flue gas is irradiated with 300–800 kV electrons to generate ions and radicals that simultaneously convert NO_X to nitric acid and SO_2 to sulfuric acid (Mätzing et al., 1988; Mätzing, 1991). The addition of ammonia gives rise to an aerosol that can be filtered in a dry state; the reaction product is a mixture of ammonium sulfate and ammonium nitrate, which is a high-quality fertilizer (Mätzing et al., 1988; Jordan, 1990).

s0110 *22.4.5 Noteworthy technologies*

p0590 SO_2 removal from humid air has been demonstrated using a pulsed steamer corona (Mizuno et al., 1986). Desulfurization using membrane technologies is promising, with polymeric membranes attracting much attention (Kim et al., 2015). The theory and science behind the membrane technology is simple; however, the practical application is complex.

p0595 The use of metal oxides has also caught considerable attention as FGD and deNOx technologies. Alumina-supported cupric oxide has the potential for use as sorbent/catalyst to remove SO_2, wherein copper acts as a sorbent by forming $CuSO_4$, and for the selective catalytic reaction of NO_X (with $CuSO_4$ acting as a catalyst) (Lowell et al., 1971; Mochida et al., 2000).

p0600 Regenerable zeolite adsorbents provide another option for removing SO_X from flue gases. A titanium dioxide (TiO_2) photocatalysis mechanism has been used to remove SO_2 and NO (98% and 67%, respectively) from flue gases (Zhao et al., 2008). In the process, SO_2 was partly oxidized to SO_3, whereas the bulk of NO was oxidized to NO_2, and both were removed by resorbing (Zhao et al., 2008).

s0115 22.5 Mercury control

p0605 Mercury (Hg) is one of the most highly volatile metals (along with arsenic and selenium) in the flue gas, and remains mostly in the gas phase. Flue-gas Hg is classified

as elemental (Hg^0), oxidized (Hg^{2+}), or particulate-bound (Hg_p). Hg_p represents only a small amount of the flue-gas Hg and is easily removed in the ESP, by an FF or using a wet scrubber. Hg^{2+} is soluble in water and is easily captured in a wet scrubber or FF. Hg^0 is the most difficult to remove, owing to its extremely low concentration in the flue gas, high volatility, low solubility, and less reactive nature.

s0120 22.5.1 Mercury emission effects

p0610 The potential of Hg for bioaccumulation in terrestrial and aquatic ecosystems has strongly driven the need for reducing its emissions from power plants. Its capacity to enter and cycle through the food chain poses a threat to public health.

p0615 A PCC plant's operations are not known to be affected by Hg. However, the presence of Hg in the degraded amine waste may make it hazardous (Sexton et al., 2014), necessitating expensive treatment procedures for safe disposal. To avoid this cost, minimization of Hg in the flue gas would be a necessary step. Hg also has amalgamating properties that may affect a PCC plant's operations and ensuing CO_2 compression/liquefaction and transport.

s0125 22.5.2 Mercury control technologies

p0620 Current Hg control technologies inject adsorbents into the flue gas to either physically or chemically bind Hg to the surface of the sorbent. The sorbent is injected before the ESP/FF to facilitate the removal of mercury in the particulate control technology (Adams, 2010). The most commonly used sorbents are fly ash, unburnt carbon in the ash, and activated carbon (Wilcox et al., 2012). The oxidation of Hg by a sorbent increases its removal rate by the particulate removal technology, because Hg^{2+} is "stickier" and more soluble (Sloss, 2015).

p0625 The capacity of any APCD that additionally removes Hg is termed a "co-benefit." Technologies offering cobenefits are ESP, FF, SCR, and FGD technologies. Research into the speciation of Hg in Australian power plants shows that around 58% of the Hg emitted in the flue gas is Hg^0 (Nelson et al., 2008), which passes through most flue-gas cleaning systems (Nicol, 2013). On average, nearly 40% of the total Hg is caught in flue-gas cleaning systems that contain some form of particulate control and a wet scrubber (Nicol, 2013), which translates to average emission range of 1 to 10 μg/m^3. In a coal-fired power plant with only a particulate control device, Hg emissions increase to 2−27 μg/m^3. An SCR system oxidizes Hg, which is soluble in water and can be removed by water scrubbing (eg, in a wet FGD system). An overview of the removal efficiencies of these technologies is provided Table 22.6.

p0630 As discussed, FFs can effectively control particulate emission via a sorbent. In all particulate control technologies, mercury removal efficiency increases at lower temperatures, due to the Hg condensing (Meij, 1997). FGD also partially removes Hg, but mostly Hg^{2+}; Hg^0 remains largely unaffected.

p0635 Aqueous solutions of sodium chloride (NaCl) and sodium hypochlorite (NaOCl) can effectively remove Hg, and can be used by including an NaCl/NaOCl absorption section upstream of the CO_2 capture process (Azzi et al., 2013).

t0035 **Table 22.6 Average mercury emission reduction for pulverized-coal-fired boilers (%) (Tavoulareas and Jozewicz, 2005)**

	Bituminous	Subbituminous	Lignite
Cold-side electrostatic precipitator (ESP)	35	3	0
Hot-side ESP	14	12	n.a.
Fabric filter (FF)	89	73	n.a.
Particulate scrubber	12	0	33
ESP and spray dry absorber (SDA)	n.a.	50	n.a.
FF and SDA	98	23	17
FF, SDA, and selective catalytic reduction (SCR)	97	n.a.	n.a.
Cold-side ESP and wet flue gas desulfurization (FGD)	81	30	42
Hot-side ESP and wet FGD	45	25	n.a.
FF and wet FGD	97	n.a.	n.a.

[AU18] *n.a.*, not evaluated.

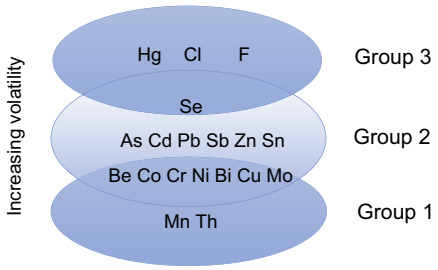
s0130 22.6 Trace elements and other contaminants

p0640 Efficient particulate control devices capture the majority of trace elements in the flue gas. Using a combination of APCDs along with injecting sorbents into the flue gas further improves capture efficiency, as shown in Table 22.7.

p0645 Fig. 22.5 classifies the main trace elements in coal according to their volatility. Group 1 elements are essentially nonvolatile, and preferentially concentrate in the bottom ash. Group 2 elements tend to concentrate in the fly ash in the form of particulates. They can also precipitate on micron-sized fine particulates when they condensate in the

t0040 **Table 22.7 Combined removal efficiency of wet flue-gas desulfurization and particulate matter control devices (Zevenhoven and Kilpinen, 2001)**

	Removal efficiency (%)	Outlet concentration ($\mu g/m^3$)
Mercury	50	~1.5
Selenium	60	~10
Beryllium	80	~250
Fly ash	90–99	1000–10,000



f0030 **Figure 22.5** Classification of trace elements by volatility during combustion (Clarke and Sloss, 1992).

cooler parts of the boiler after their initial volatilization. Group 3 elements, which are highly volatile and stay in a gaseous form, are emissions of concern in both coal-fired power plants and PCC plants.

p0650 The tendency of some elements (Cr, Ni, As, Se, Cd, Sb, Hg, and Pb) to be enriched in the submicron fraction of fly ash (Davidson and Clarke, 1996) helps them pass through any particulate control technology. This fine ash fraction would inevitably enter the PCC plant. The chemical composition of such fine particulates depends on the concentrations of specific elements. For example, if calcium concentration is high enough, then arsenic and selenium may form calcium arsenate and selenate.

s0135 22.7 Multipollutant control

p0655 A multipollutant control system is defined as a process that removes two or more of the principal regulated pollutants using a single reactor or technology. SO_2, NO_X, PM, Hg, and CO_2 are the targeted pollutants in this regard.

p0660 The lower capital and operating costs are the main attraction for multipollutant control technologies. The smaller footprint and maximization of marketable products is another area of focus in these systems, with their smaller footprint making them an attractive option for retrofit.

s0140 *22.7.1 Multipollutant control (CO_2 inclusive)*

s0145 *22.7.1.1 Cansolv® technology*

p0665 The Cansolv® technology of Shell (Shaw, 2009) is an integrated SO_2- and CO_2-removal system. SO_2 is scrubbed from the flue gases followed by CO_2 capture. The system uses a proprietary aqueous amine solution for 99.9% removal of SO_2, resulting in an optimized slip of SO_2 into the CO_2 capture system. This is followed by CO_2 removal, achieving 90% efficiency of capture of CO_2 and capturing the remainder of the SO_2. The SO_2 and CO_2 loops are completely separate in regard to mass flows and regeneration technology, but are integrated in regard to optimizing heat transfer. As the system removes around 50% of the SO_3, a separate system is needed to reduce its concentration to an acceptable level; such a system has been installed in the

Boundary Dam carbon capture project. Cansolv's water consumption is lower than other wet scrubber systems, mainly because it is a regenerable system. SO_2 is converted into sulfuric acid, which can be sold as a product. The process takes SO_2 levels of >2000 ppm, and developments are under way to scrub newly designed sour gas-fired power stations, in which SO_2 levels of >10,000 ppm are expected.

s0150 *22.7.1.2 CASPER and CS-Cap processes*

p0670 The CASPER process resulted from collaboration between European and Australian industry and research organizations. The Europeans were aiming to modify their existing SO_2 capture systems and capture as much as possible CO_2 simultaneously. This was combined with Australian efforts to combine SO_2 and CO_2 capture in one single unit, because no FGD plants were installed in Australia due to the country's low-sulfur coals.

p0675 [**AU10**] The Netherlands Organization for Applied Scientific Research (TNO) and Commonwealth Science and Industrial Research Organization (CSIRO) collaboratively developed the CASPER process, which was proven at pilot scale in which SO_2 and CO_2 were captured into a model amino acid, beta-alanine, in one single loop (Heffernan et al., 2014). The lowest solubility of potassium sulfate was in the cold, lean liquid absorbent. Once loaded with sulfate, this stream was cooled and potassium sulfate was removed through crystallization. The resulting blend, which contained the amino acid, was utilized for capture of CO_2 and SO_2 in a cyclic way.

p0680 The main advantage of the CASPER process is that additional FGD is not required. The large piece of equipment required to process large volumes of flue gas prior to the PCC process can be replaced with a small crystallization unit, which processes a slipstream of the liquid absorbent working in the PCC process.

p0685 CSIRO's CS-Cap (Meuleman et al., 2012) process also combines SO_2 and CO_2 capture into a single process, thereby significantly reducing capital and operating costs. This process also uses one liquid absorbent; however, it operates in two separate loops, capturing the SO_2 and CO_2 at different pH settings. The advantage of this process is that SO_2 is captured continuously at optimal settings, and is tolerant to significant variations in SO_2 concentration in the flue gas from the power plant due to combustion of variable sulfur-containing coals. Regenerated liquid absorbent from the SO_2 loop is fed back to the CO_2 capture loop; in turn, rich absorbent from the CO_2 capture loop is fed at the top of the SO_2 capture section to control the pH and thereby the SO_2 breakthrough concentration. To counter the variable SO_2 rates the process can simply vary the rate of regeneration of the absorbent in the SO_2 capture loop.

p0690 [**AU11**] Both technologies can capture all the SO_2 from the flue gas (up to 700), whereas the SO_2 concentration in the emitting stream is well below 5 ppm (Cousins et al., 2015).

s0155 *22.7.1.3 CO2CRC UNO MK 3 process*

p0695 [**AU12**] The CO_2 Cooperative Research Center (CO2CRC)'s UNO MK 3 (Hooper, 2015) is a precipitating aqueous carbonate process system that uses potassium carbonate (K_2CO_3) to remove CO_2 from flue gases. The process also captures SO_X and NO_X to produce potassium sulfate (K_2SO_4) and nitrate (KNO_3) salts. A pilot plant at

Hazelwood (CO2CRC/GDF-SUEZ) in Australia was commissioned in 2012. Further process development includes a search for applicable accelerators to increase the kinetics of the capture process.

s0160 *22.7.1.4 Chilled and cooled ammonia processes*

p0700 Chilled ammonia, operating at subambient temperatures, also has the potential for multicomponent removal. The best-known technology in this area is the chilled ammonia process developed by Alstom, who have completed a demonstration project for CO_2 capture from flue gas. The system has high CO_2 capture rates (up to 87%) with a CO_2 purity of 99.9% and low NH_3 emissions (Lombardo et al., 2014). A similar technology operated at a near ambient temperature is the cooled ammonia process as developed by CSIRO.

p0705 There are a number of potential benefits of an ammonia-based process. Being the most fundamental amine, ammonia is the cheapest capture absorbent to produce and very robust as it does not degrade. Being robust, it can operate at higher stripper temperatures and pressures that can produce higher-pressure CO_2 stream, which can reduce the number of expensive compression stages required for CO_2 liquefaction. Being a cheap liquid absorbent, it can be readily consumed to capture SO_2 and NO_2. In fact, it is possible that salable byproducts of ammonium sulfate and ammonium nitrate could be produced with the right process combination.

p0710 Ammonia is a volatile compound, however, and introduces some difficulties in terms of treatment and limiting slip, hence the general requirement for operating at lower temperatures, which increases cost of operation. Ammonium bicarbonate can also precipitate in the system, and if the system is not designed to handle or avoid this precipitation, it can cause problems. Alstom's chilled ammonia process was designed to handle some precipitation.

s0165 *22.7.1.5 Carbon fiber composite*

p0715 A carbon fiber-composite solid sorbent has been tested at a coal-fired power station in New South Wales, Australia. The prototype consistently achieved very high levels of removal of SO_2, NO_X, and CO_2 from flue gas (Thiruvenkatachari et al., 2015). The design of the module allows for low-pressure drop of the flue gas, while providing high surface areas for adsorption. Although the system was developed for CO_2 capture, the results have shown that it should be considered for future multicomponent pretreatment systems.

s0170 *22.7.2 Multipollutant control (exclusive of CO_2)*

p0720 Several noteworthy technologies remove multiple impurities, but are beyond the scope of this chapter. These include:

u0170 • Ammonia-free SCR (Fan et al., 2005)
u0175 • Greenidge Multi-Pollutant Control Project (Connell et al., 2007)
u0180 • Indigo Multi Air Pollutant System (MAPSystem™) (Carpenter, 2013)
u0185 • Regenerative Activated Coke Technology (ReACT™) (Dene et al., 2008)

u0190 • ElectroCatalytic Oxidation™ (Schimmoller, 2003)
u0195 • Haldor Topsoe (Denmark) SNOX™ (Carpenter, 2013)
u0200 • Dynawave Membrane System (Nicol, 2013)
u0205 • Pulsed streamer corona (Mizuno et al., 1986; Clements et al., 1989).

p0765 The aforementioned technologies have inherent strengths and limitations. The ReACT™ process has lower NO_X removal efficiency than SNOX™. Both are dry and regenerable processes. ReACT™ is better suited for low to medium sulfur coals, whereas SNOX™ is economically attractive for plants firing high-sulfur coals (Carpenter, 2013). The Electro-Catalytic Oxidation (ECO®) technology, while removing NO_X, SO_2, SO_3, HCl, and other acid gases, requires the SO_2 to NO_X molar ratio of three or more for optimum performance (Carpenter, 2013). High-sodium coals pose operational issues for ECO® technology as well (Carpenter, 2013). The AF-SCR system provides large cost savings over the conventional SCR system (Nalbandian, 2009). When activated carbon catalyst is used, it acts as a sorbent to capture/convert SO_X chemically. The SO_X-adsorbed activated carbon is then used to efficiently capture mercury. Work continues on the AF-SCR system development with financial support from the US Department of Energy [AU13] National Energy Technology Laboratory (DOE NETL) (Nalbandian, 2009). Greenidge [AU14] MultiPollutant Control Project (Connell et al., 2007) is currently being demonstrated at the 107 MWe AES Greenidge unit 4 in Dresden, New York, United States. The Dynawave Membrane System is based on Wet ESP technology. It combines limestone slurry scrubbing and membrane ESP into one unit with the capacity to remove SO_3, SO_2, oxidized mercury, and fly ash. Pulsed Streamer Corona has the potential for multiproduct removal including SO_2, NO, and fly ash; however, the research is still in the initial phases.

s0175 22.8 Conclusion

p0770 As a result of coal combustion, flue gas contains constituents that include most of the elements of the periodic table. Flue-gas impurities such as particulate matter (PM), sulfur oxides (SO_X), and nitrogen oxides (NO_X) are commonly removed through technologies that have been integrated with power plant operation since the previous century. The removal of others has only recently begun to be addressed.

p0775 Most pollution control strategies have addressed environmental impacts through related legislation or permitting requirements. Other strategies have been implemented simply because they were beneficial to the economic operation of the power plant, either through increased efficiency or lower maintenance requirements. Of key interest in this chapter were the implications of the sensible removal of impurities to optimize the operation of postcombustion capture of carbon dioxide (PCC).

p0780 DeNOx and PM removal technologies are close to the specifications required by PCC, but improvements would be beneficial. In contrast, the most economic flue-gas desulfurization (FGD) technologies, which are based on limestone, are often unsatisfactory. They comply with SO_2 emission legislation, directives, and permits; however, the resulting concentrations of 30 to 70 ppm are too high for most PCC technologies. After flue gas has been treated in a PCC unit, the SO_2 levels in the treated gas

reduce to below 1 ppm. This difference in SO_2 concentrations is accounted for by absorption into the liquid absorbent, which consequently deteriorates its efficiency as a CO_2 capture agent through lowering the pH and the formation of heat-stable salts.

p0785 Many technologies address a single-component issue. By placing treatment units in series, a multicomponent removal train can be operated in conjunction with power generation using coal. However, most technologies that address one issue simultaneously contribute to worsening another issue. Therefore, logical placement of separation units is paramount.

p0790 The addition of the PCC process is creating further optimization challenges that must be solved holistically. For example, the combination of fly ash and corrosion will accelerate blockage of critical process unit elements, such as heat-exchanger internals and column packing material. Technologies that reduce SO_3 concentrations are strongly preferred, because SO_3 is notorious for creating aerosols that cannot be caught with low-pressure drop units, leading to high liquid-absorbent losses. Corrosion is another optimization issue, which can be induced by HCl, or accelerated by sulfate or nitrates, heat-stable salts, or degradation products of the original liquid absorbent.

p0795 Some new systems have addressed the combined requirement on emissions. The most advanced of these is Shell's Cansolv technology, which operates at commercial scale generating 120 MW at Saskpower, Canada, since October 2014, while removing PM, NO_X, SO_X, and CO_2 by at least 90%. Other technologies focusing on multicomponent flue-gas treatment of impurities (with or without CO_2 capture) are under development by various research organizations, companies, and consortia.

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[**AU17**]

FERON: 22

Non-Print Items

Abstract

[AU2] This chapter discusses the importance of flue-gas treatment and the effect of its impurities on postcombustion CO_2 capture (PCC) process performance. Important consequences of nonoptimized flue-gas treatment include atmospheric emissions, amine degradation, extra maintenance requirements through corrosion or fly ash deposition, and waste handling. Each of these areas is strongly dependent on the others. We briefly describe existing flue-gas separation technologies and compare them to the requirements of PCC. Further pretreatment technologies are suggested to improve the composition and thereby the properties of the flue gas entering the PCC plant. For optimal plant operation, the overall process from boiler to atmosphere and waste needs to be analyzed holistically, because:

- certain approaches can increase efficiency when treating multiple contaminants compared with treating each component separately;
- most "cleaning" steps influence or generate other compounds that could reduce the performance of the process chain;
- all costs associated with the treatment chain increase production costs, with little or no increase in value.

Keywords:
Air pollution control devices, Flue-gas separation, Flue-gas treatment, Fly ash deposition, Impurities, Nitrogen oxides, Postcombustion CO_2 capture, Sulfur oxides.